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ARTICLE

Control of Primary Silicon Crystal Size in Hypereutectic Al-Si Alloys by Filtering Casting Method

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Abstract: A main technical problem in the preparation of hypereutectic Al-high Si alloy is the refinement of the primary Si particles. In this paper, a new method for the preparation of Al-high Si alloy with refined primary Si particles by filtration treatment was proposed. During this process, the melt of hypereutectic Al-Si alloy underwent the filtration treatment at the temperature little above the eutectic line in the Al-Si binary phase diagram. Afterwards, the large-sized primary Si particles were retained by the filter screen, while the small-sized primary Si particles together with the eutectic melt went through the screen and cooled down, forming the hypereutectic Al-high Si alloy products. The results show that the Si content in the Al-high Si alloy prepared in this study is about 27 wt%, the average diameter of the primary Si particles is less than 45 μm, the roundness is 1.43, and the Brinell hardness (HB) of the alloy is about 700 MPa. By this method, it is expected to obtain the Al-high Si alloy with higher degree of sphericity of the primary Si particles and higher Si content.

Key words: hypereutectic Al-Si alloys; refinement; primary Si; filtering; Al-Si

Due to the presence of primary Si particles in the alloy microstructure, the hypereutectic Al-high Si alloys (Si content is above 16 wt%) are characterized by high wear resistance[1, 2], high specific strength, low thermal expansion and excellent castability[3]. As a consequence, they are applied extensively in aerospace, automobile manufacturing, electronic devices and other fields. The advantages such as high wear resistance, low thermal expansion coefficient and low density can help to produce long life, lightweight combustion engine components. And when the hypereutectic Al-high Si alloys used utilized as the structural materials in aircrafts or automobiles, the weight of products can be greatly reduced due to the low density and high specific mechanical strength.

With the continuous improvement of engine performance, requirements for the Al-Si alloy on the thermal expansion coefficient, wear resistance and other performances are getting higher and higher. Thus, the Si content in the Al-Si alloy should be further increased. The increase of Si content will improve the high temperature strength of the alloy and reduce the thermal expansion coefficient. However, with the increase of the Si content in the hypereutectic alloy, the crystallization temperature range of the primary Si phase becomes wider and the crystallized primary Si particles becomes very large, resulting in the high brittleness[4], poor casting performance and machinability. Under conventional casting conditions, if the primary Si crystal phase is not refined, it is difficult to obtain qualified castings when the silicon content exceeds 20 wt%. This is the main reason that restricts the application of Al-high Si alloys.

In order to solve above defects, the only way is to refine the primary Si particles[5, 6]. It is found that the smaller size, better roundness and more uniform distribution of primary Si particles can contribute to higher tensile strength and better overall performance of the Al-Si alloy[2, 7, 8]. While the irregular primary Si particles will seriously dissever the aluminum matrix when suffering high mechanical impact[9]. At present, the most widely used method for refining primary Si crystal grains is adding, etc. master alloy modifiers[5, 10, 11] such as Cu-P, Al-Cu-P into the alloy. But its refining effect is usually limited and incontrollable in some circumstance. The primary Si grain

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refining effect will be significantly weakened, in particular when the Si content is above 18 wt% in the Al-high Si alloy. In addition, the agitation casting and rapid solidification methods[12-14] have also attracted attention from many researchers. For instance, the introduction of agitation casting[15] on the basis of the existing casting process can contribute to better refinement effect of primary Si crystals; and the rapid solidification method such as the jet deposition can be used to obtain the products with fine primary Si grains and uniform structure[14]. However, above methods call strict requirement in equipment as well as complicated operation. However, the high cost of spray deposition method, leads to its barrier for large-scale production. The traditional methods can only achieve the grain refinement of primary Si statistically, while cannot limit the maximum grain size of primary Si.

In order to restrict the size of primary Si particles directly and effectively, and to make influence on the casting process as little as possible, the filtering casting method is proposed in this study. By the pressure filtration method the large-sized primary Si particles and the Al-Si melt are physically separated. When large-sized primary Si particles are intercepted by the screen and the small-sized primary Si particles together with the eutectic melt flow through the screen, then cool down and solidify in the mold, finally the Al-high Si alloy with fine primary Si particles is obtained. By this method, the regulation of the maximum primary Si particle size in the alloy can be realized. In this study, the average Si content in the Al-high Si alloy has reached 27.1 wt%, the average diameter of the primary Si particles has reached less than 44.5 μm, and the average roundness of the particles has reached 1.43. The method has the advantages of simple operation, easy control, obvious effect of primary Si particle refinement and good particle size uniformity.

1 Experiment

1.1 Materials

The raw materials used in the experiment are metallurgical grade silicon (MG-Si) numbered 3303 and high purity aluminum, both of which compositions are shown in Table 1. Their physical photos are shown in Fig.1. Large pieces of MG-Si need to be crushed before utilization, while short rods of high purity aluminum can be used directly.

1.2 Apparatus

As shown in Fig.2, the filtering casting apparatus used in the experiment is mainly composed of two chambers, and the upper high pressure chamber is equipped with induction coil for heating the graphite crucible and the internal Al-Si alloy sample. A platinum and rhodium thermocouple (type S) was used for the sample's temperature determination. The interspaces between the chamber and the induction coil and also between the induction coil and the graphite crucible are filled with fire-resistant thermal insulation material. The structure of the graphite crucible is shown in Fig.2, consisting of three parts, a sample containing part, a middle porous support, and a lower seal support. In the experiment, above the porous graphite sheet was padded with stainless steel screen and graphite felt, which was used to filter and separate the primary Si particles of different sizes. The cooling water jacket between the upper and lower chambers and the graphite seal support were sealed by an inclined surface, so as to ensure that the alloy melt in the graphite crucible could finish the filtering process under the high pressure of the upper chamber. And the filtrate flowed into the graphite crucible in the lower chamber and then cooled and solidified. The metal walls of the entire apparatus were cooled by circulating water.

1.3 Procedures

The experimental procedures are illustrated in Fig.3, which can be divided into four parts as follows.

1.3.1 Induction furnace pre-melting

MG-Si and high purity aluminum were pre-melted in a vacuum induction furnace at the ratio of Si 45 wt% and Al 55 wt% using an alumina crucible as container. The maximum pre-melting temperature was 1873 K. Then the completely alloyed Al-Si melt cooled with the furnace. Finally, after removing the alumina crucible, the pre-melted hypereutectic Al-Si alloy was obtained.

1.3.2 Wire-electrode cutting

The hypereutectic Al-Si alloy bulk prepared as described above was cut by a wire cut electric discharge machine. Several columnar alloys with the size of Φ24.5 mm×30 mm and the mass of about 30 g were obtained.

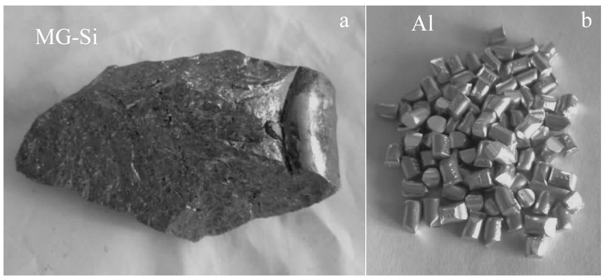

Fig.1 Pictures of MG-Si (a) and high purity Al (b)

Table 1 Compositions of the MG-Si and high purity Al (μg/g)

Element	Ca	Ti	V	Fe	Al	Mg	Si	Cr	Mn	Ni	Cu	B	P
MG-Si	1183.21	326.26	533.26	3922.60	438.10	143.38	Bal.	234.83	171.57	104.41	41.31	38.87	97.57
Al	38.4	2.85	11.42	311.56	Bal.	20.95	-	3.47	3.23	6.4	2.28	4.31	7.51

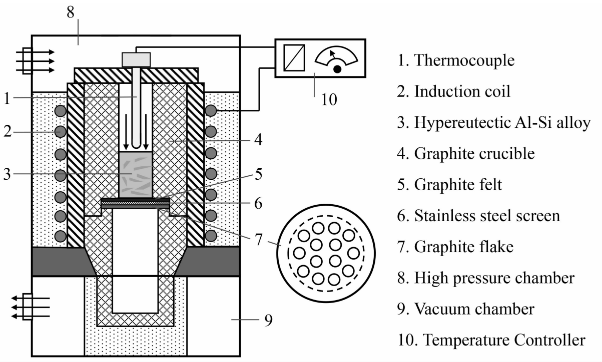

Fig.2 Sketch map of the filtering casting apparatus

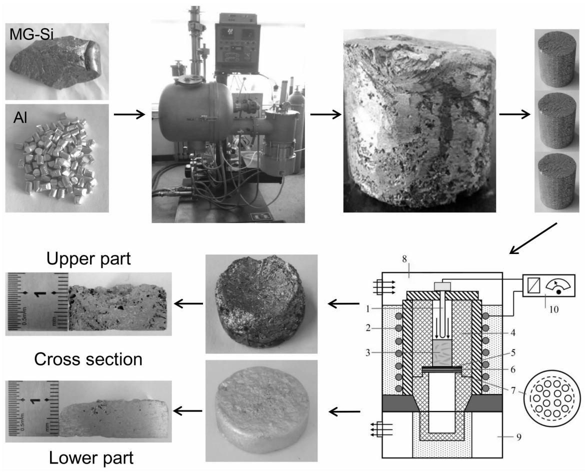

Fig.3 Sketch map of the experimental procedures

1.3.3 Pressure filtration

Firstly, the graphite crucible for pressure filtration was assembled. As shown in Fig.2, the filter part includes three layers, from top to bottom which are the graphite felt of 1 mm in thickness, stainless steel screen (the mesh sizes used in the experiment are 250, 150, 75, 38 μm) and graphite flake with several Φ2 mm holes. Then the columnar alloy was put into the graphite crucible, and the graphite crucible was put into the high pressure chamber and tightly combined with the cooling water jacket to ensure sealing. Then the thermocouple was installed, and the high pressure chamber cover was closed. After 2~3 times evacuation-Ar gas charging operation, the heating program was started. The Al-Si alloy was heated to 1373 K and melted, after which it was cooled at a rate of 3 K/min to 973 K and kept for 1 h. The temperature profile is shown in the Al-Si binary phase diagram is shown in Fig.4. After turning on the gas tank switch, the Ar gas instantly filled into the high pressure chamber, and the pressure was maintained at (0.1 ± 0.02) MPa. When the filtration was completed, the pressure in the high pressure chamber gradually decreased. Then the heating power was turned off, so that the sample cooled with the furnace. After the furnace temperature dropped to room temperature, the samples were released from the two chambers, as shown in Fig.3.

1.3.4 Samples preparation and tests

The upper part on the filter and the lower part refined alloy were cut along the longitudinal section, half of which was used to measure the silicon content and the other half was firstly ground with SiC abrasive paper, then polished using 1 μm diamond suspension and finally observed under an optical microscope (DM4M, Leica, Germany). The Image Pro 6.0 software was used to quantify the size of the primary Si particles in the refined Al-Si alloys. The mean diameter, maximum diameter, minimum diameter, solid fraction and roundness values of the primary silicon particles can all be calculated by the software. The proportion of the area occupied by the primary Si particles in a picture was defined as the solid phase fraction. The roundness was used to measure the sphericity of the primary Si particles. The smaller the value was, the higher the sphericity was. The mass fraction of Si in the alloy was determined by a chemical method: firstly the aluminum phase is dissolved with hydrochloric acid, and then the Si phase in the residual can be got after filtering and drying. The hardness tests were performed by a Brinell hardness tester (THBS-3000MDX, China) equipped with a carbide ball, 10 mm in diameter, and a load of 2500 N was applied for 10 s.

2 Results and Discussion

2.1 Separation effect of the filtering process

The samples mass of the upper and lower parts in the experiment is shown in Fig.5. It can be seen that the mass ratio between the two parts is relatively constant. The weight of the upper residual part is about 10 g and that of the lower refined Al-Si alloy part is about 20 g. In general, the amount of loss in the experiment is very little, only about 1wt%.

Fig.6 shows the Si content in the residual part and refined Al-Si alloy as well as the Si recovery rate in the experiments with different mesh sizes. The Si recovery rate is defined as

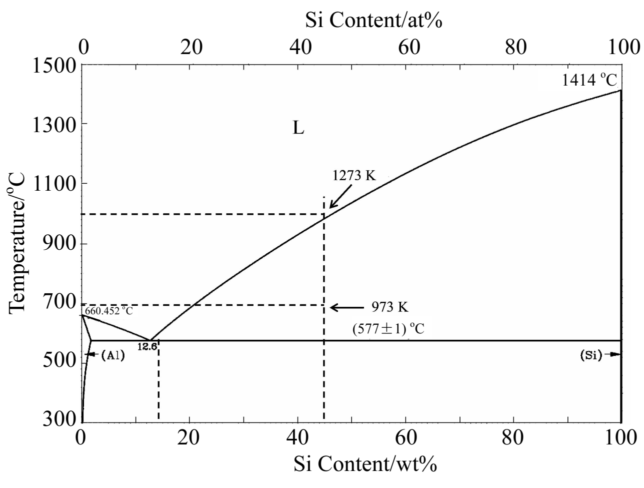

Fig.4 Temperature profile during pressure filtration process

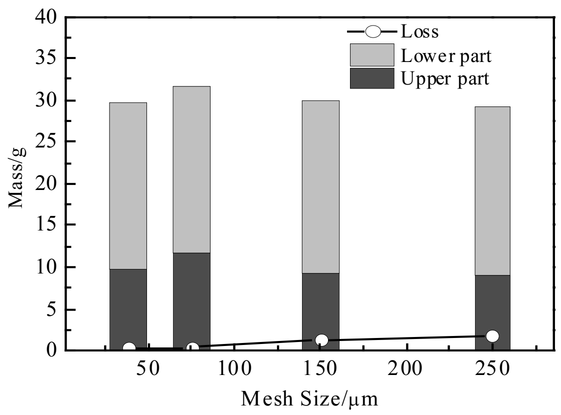

Fig.5 Samples mass of the upper and lower parts with different mesh sizes

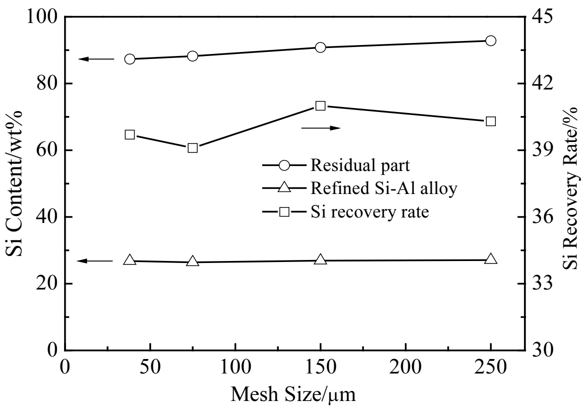

Fig.6 Si content and recovery rate versus mesh size

the percentage of Si in the refined Al-Si alloy accounting for the total Si in the raw material. It can be seen that the Si contents in the residual part and the refined Al-Si alloy basically are maintained at 90 wt% and 26 wt%. So it is proved that the Si content in the Al-high Si alloy does not change a lot with the mesh size, and it can be kept very stable with this filtering casting method. The Si recovery rate is about 40% regardless different mesh sizes.

2.2 Refining effect of primary Si particles in alloys

2.2.1 Effect of mesh size on the size of primary Si particles

The microstructure of the Al-high Si alloy obtained by the filtering casting method with different mesh sizes is shown in Fig.7. It can be seen that with the decrease of the mesh size, the size of the primary Si particles in the Al-Si alloy significantly reduces. And the shape of the primary Si particles gradually changes from the slat shape to the equiaxed shape. Meanwhile, the halos around the primary Si particles become more and more apparent, and the number of spherical α-Al particles increases. However, the dendrite α-Al phase is not found in the alloy. For the eutectic phase, the elongated β-Si phase in it gradually changes from coarse and long to thin and short.

The size distribution of the primary Si particles calculated with Image pro 6.0 software is shown in Fig.8. It can be seen that the mean diameters of the primary Si particles in the alloy after filtration with the mesh size of 250, 150, 75 and 38 μm are about 125, 75, 65 and 45 μm, respectively. Fig.9 shows the relationship between the mesh sizes and the maximum, the minimum, the average particle size of the primary Si. The maximum diameter of the primary Si particles is larger than that of the corresponding mesh size, and with the decrease of the mesh size, the gap between the maximum particle diameter and the corre-

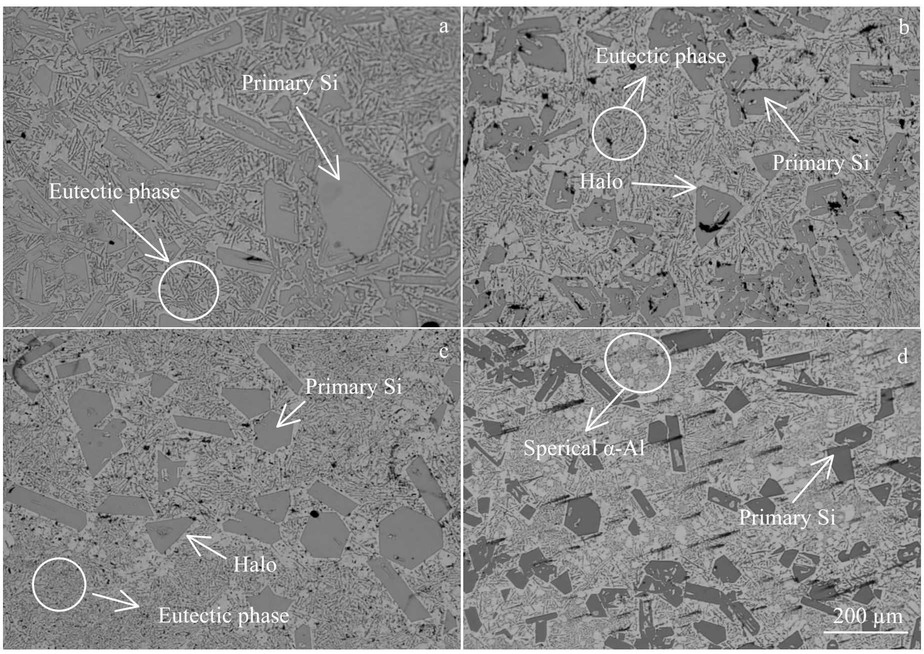

Fig.7 Metallographs of the refined Al-Si alloy prepared with different mesh sizes: (a) 250 μm, (b) 150 μm, (c) 75 μm, and (d) 38 μm

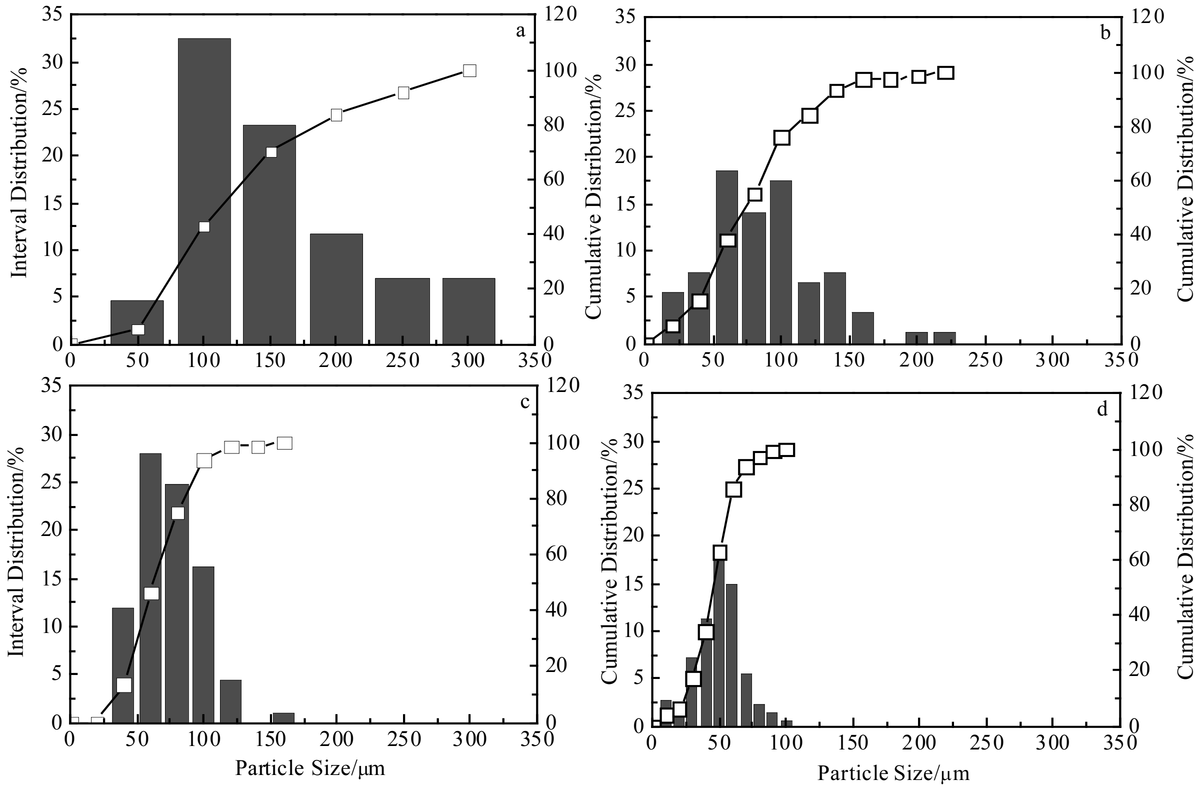

Fig.8 Particle size distribution of primary Si with different mesh sizes: (a) 250 μm, (b) 150 μm, (c) 75 μm, and (d) 38 μm

sponding mesh size increases. On the one hand, it is attributed to the rod-like shape of the primary Si particles, as some of the larger size primary Si particles may be filtered through the mesh along the direction of the smallest cross-section. In addition, excluding the solid primary Si particles, the alloy mother liquor at 973 K before filtration is still hypereutectic, as shown in Fig.4. When the small-sized primary Si particles are filtered together with the eutectic mother liquor into the cold mold, the excess Si in the eutectic melt is likely to promote the secondary growth of the primary Si in the alloy to promote the generation of new primary Si particles until the hypereutectic melt becomes a eutectic system. In order to further reduce the size of the primary silicon particles in the alloy, we can further reduce the mesh size under the premise of not affecting the filtration effect. In addition, we can continue to lower the temperature of the melt and make it closer to the eutectic temperature (850 K) before filtering, which will reduce the hypereutectic degree of the melt and avoid the secondary growth of the primary Si particles in the alloy after filtration. Unlike the wide difference of the primary Si particles' maximum diameters in the experiments with different mesh sizes, the minimum diameters just change within the range of 14~44 μm, which are not sensitive to the mesh size. The mean diameter in Fig.9 is very close to the median value of the particle diameter in Fig.8.

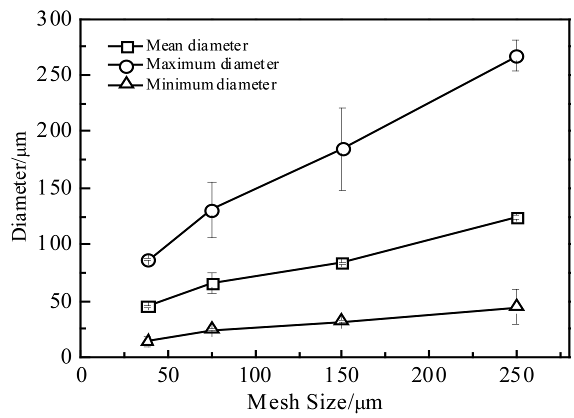

Fig.9 Relationship of the mean, maximum and minimum diameters to mesh size

2.2.2 Influence of mesh size on the shape of the primary Si particles

Fig.10 illustrates relationship of the solid fraction and roundness of the primary Si particles with the mesh sizes. They all increase with the increase of the mesh size. It proves that with the decrease of the mesh size, the primary Si particles in the Al-Si alloy are not only refined in size but also equiaxed in shape. This phenomenon is also revealed in Fig.7: when the mesh sizes

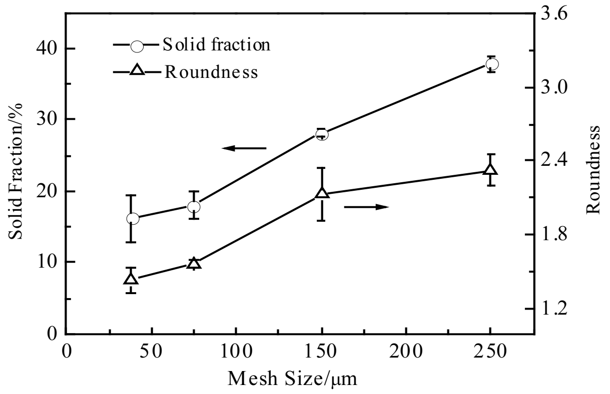

Fig.10 Relationship of the solid fraction and roundness to mesh size

are 250 and 150 μm, the primary Si particles are usually in the shape of strip or gathered coarse bulk, which will cause the low ductility of hypereutectic Al-Si alloys.

The primary Si particles present the equiaxial polygon shape or short rod-like shape when the mesh sizes are 75 and 38 μm. It is found in some research that the mechanical properties of hypereutectic Al-Si alloys largely depend on the size, morphology and distribution of the primary Si particles[16-19]. Thus, the regulation on the size and shape of the primary Si particles with this method can contribute to the improvement of mechanical properties of the Al-high Si alloy.

2.2.3 Brinell hardness test of the as-prepared Al-Si alloy

The Brinell hardness (HB) of the as-prepared Al-Si alloy with different mesh sizes makes little difference, which is about 700 MPa. Generally, higher Si content contributes to higher hardness of the Al-Si alloy. The average Brinell hardness of the upper part above the filter in this experiment is about 2010 MPa

2.3 Contrast experiment without filtration treatment

In order to prove the refinement effect of the primary Si with the filtering method, some contrast experiments without filtering treatment were carried out. Firstly, the MG-Si and pure Al were mixed with the mass ratio of 27:73. To figure out the primary Si refinement effect with phosphorus based master alloys, 1.4 wt% of Cu-P master alloy was added into two samples. Then those samples were put into graphite crucibles with lids and melted in a muffle furnace at 1173 K for 1 h. Finally, they were cooled with the furnace (slow cooling) or water quenched (rapid cooling). The cross section morphologies under optical microscope of the as prepared Al-27%Si alloys with different experimental conditions are shown in Fig.11.

It can be seen that is added Cu-P master alloy or not, the primary Si crystals will grow into big bulks. This is caused by the prolonging of cooling time and provides plenty of time for the growth of the primary Si particles. Moreover, the primary Si particles and the β-Si particles within the eutectic phase can be refined obviously under water cooling conditions, and the refinement effect of the primary Si particles is better with adding 1.4 wt% of Cu-P. While the refinement effects of the primary Si particles in the contrast experiments are all inferior to those with filtering treatment when the screen size is kept under 150 μm. There are still lots of clusters of primary Si particles even under water cooling conditions and with 1.4 wt% Cu-P addition. Compared to Fig.7, it can be deduced that the filtering treatment can also help to break up those primary Si clusters into several smaller particles.

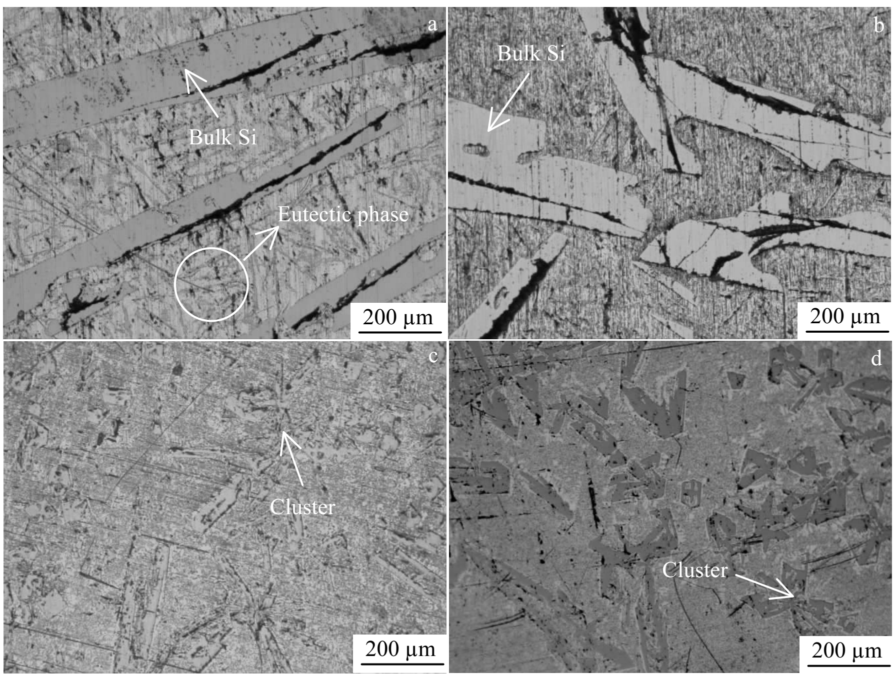

Fig.11 Metallographs of the Al-27%Si alloys prepared without filtering treatment: (a) no Cu-P slow cooling, (b) 1.4 wt% Cu-P slow cooling, (c) no Cu-P water cooling, and (d)1.4 wt% Cu-P water cooling

2.4 Design of the technological process

A conceptual combined filtering casting process for producing primary Si refined hypereutectic Al-Si alloys is proposed in this paper. The flowchart of the process is shown in Fig.12. Firstly, the mixed Si, Al and some additives for primary Si refinement, eutectic phase modification and grain refinement are pre-melted in an induction furnace. The Al-Si melt is then placed in a container with a filter screen at the bottom, and the temperature of the melt is reduced to near the eutectic temperature. After the primary Si is sufficiently precipitated, the gas pressure is applied to the vessel and the filtration takes place. The filtrate of Al-high Si alloy can be cast into alloy ingots as deep processing raw materials. In addition, with the combination of the casting process, the Al-high Si alloy melt can be directly poured into the special-shaped mold to produce net-shape Al-high Si alloy products. This process can not only prepare the Al-high Si alloy products with Si content above 30 wt%, but also can control the size of primary Si particles in alloy products, especially the maximum Si particle size can be strictly controlled. Consequently, the fracture toughness of the Al-high Si alloy can be improved, and it will help to produce the high-end aluminum alloy products with high wear resistance, high strength, and low thermal expansion coefficient.

2.5 Discussions

The performance of Al-high Si alloy is up to lots of processing parameters and conditions, and it is difficult to obtain the product with satisfying properties in every aspect with only one method. So usually combined performance optimization methods[15, 20] are performed in practical production process, such as the rapid solidification method[6, 21] for primary Si refinement and grain refinement, and adding additives as described in section 2.4. The filtering casting method as introduced in this study is proved to be a simple and effective physical method for primary Si refinement during Al-high Si alloy production. The main problems for the realization of this method are choosing a proper filter plate material and establishing a sealed filtering vessel. It can be seen in this experiment that the mass fraction of Si in the as-produced Al-high Si alloys can be kept at a certain value just with tiny fluctuation by the filtering method. The maximum size of primary Si particles that can be restricted with the filtering method is needed to be figure out in later studies.

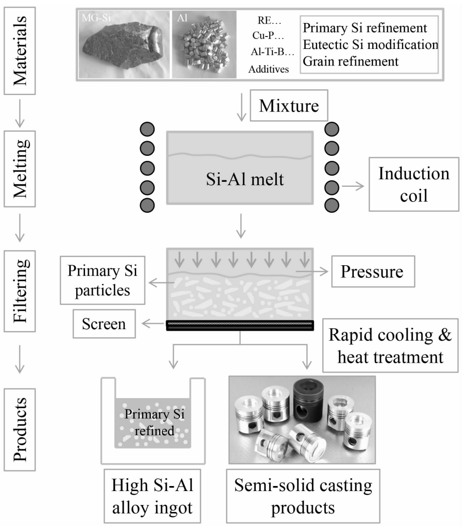

Fig.12 Schematic of the proposed filtering casting process

3 Conclusions

1) Reducing the filtering mesh size can achieve a significant effect on the refinement of the primary Si particles, and contribute to the spherical transformation of the primary Si particles.

2) Si content in the Al-high Si alloy prepared in this experiment is about 27 wt%. And the hypereutectic Al-high Si alloy of relatively stable Si content can be obtained by this method.

3) It will help to produce high quality Al-high Si alloy products if this method is integrated with the general casting process.

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过滤铸造法调控过共晶硅铝合金中初晶硅尺寸

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摘　要：初晶硅颗粒的细化是过共晶高硅铝合金制备过程中的一个主要问题。本研究提出了一种通过过滤的方式来获得含有细化初晶硅颗粒的高硅铝合金的方法。在过滤铸造的过程中，过共晶硅铝合金熔体保持在 Al-Si 二元相图共晶线偏上一点的温度。大颗粒初晶硅被筛网阻拦下来，细小的初晶硅颗粒连同共晶熔体穿过筛网进入下部坩埚冷却结晶。在本研究中通过以上方法所制备的高硅铝合金中硅的质量分数约为 27%，初晶硅颗粒的平均直径在 45 μm 以下，圆度为 1.43，布氏硬度(HB)为 700 MPa。通过该方法有望制备出初晶硅颗粒球形度更高、硅含量更高的高硅铝合金产品。

关键词：过共晶硅铝合金；细化；初晶硅；过滤；Al-Si

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